

SCRATCH AND MAR RESISTANT SOFT ETHYLENE ELASTOMER COMPOSITIONS

Field of the Invention

The invention relates to the field of soft ethylene elastomer compositions and in particular to soft ethylene elastomer compositions exhibiting improved scratch and mar resistant properties.

Background to the Invention

The manufacture of exterior automotive parts has traditionally involved hard ethylene elastomer compounds, which provide good impact and scratch and mar resistance properties to the automobile exterior. These ethylene elastomers comprise a majority of polypropylene, a lesser amount of a rubber toughener (EPDM or EPR rubber) and a filler such as talc.

More recently compounds have been developed with added softness. These compounds called "soft ethylene elastomer compounds" have a polyethylene elastomer as the major component of the composition and polypropylene as a minor component. The addition of a small amount of organic peroxide, optionally with a co-agent, changes the melt rheology characteristics of the compound thereby improving thermal resistance and processing characteristics of the compound and providing a soft characteristic to the compound.

An example of a soft ethylene elastomer compound is disclosed in U.S. Patent No. 6,506,842 and International Patent Application No. WO 98/32795 to Heck et al. published July 30, 1998, which are incorporated herein by reference. U.S. Patent No. 6,506,842 and WO 98/32795 disclose a rheology-modified, substantially gel-free ethylene elastomer composition comprising an ethylene alpha-olefin (EAO) polymer or EAO polymer blend and at least one high melting polymer selected from polypropylene homopolymers and propylene/ethylene (P/E) copolymers. The composition has at least three of the following four characteristics: a shear thinning index (STI) of at least 20, a melt strength (MS) of at least 1.5 times that of the composition without rheology modification, a solidification temperature (ST) of at least 10°C greater than that of the composition without rheology modification, and an

upper service temperature (UST) limit of at least 10°C greater than that of the composition without rheology modification.

The EAO polymer or EAO polymer blend of the ethylene elastomer is present in an amount of 50-90 % by weight of this component, preferably 65-85 % by weight while the high melting point polymer is preferably present in an amount of 50-10% by weight, more preferably 35-15% by weight.

U.S. Patent No. 6,506,842 and WO 98/32795 also disclose a process for preparing a rheology-modified, substantially gel-free thermoplastic elastomer composition, which includes treatment with an organic peroxide, and manufacture of articles using these compositions.

For automotive interior and other applications that require scratch and mar resistance, soft ethylene elastomer compositions have been deficient due to their soft polymeric nature.

As a result there still remains a need to provide a soft ethylene elastomer composition producing compounds with improved scratch and mar resistance properties.

Summary of the Invention

According to one aspect of the invention there is provided a composition comprising a soft ethylene elastomer component and an effective amount of filler to impart improved scratch and mar resistance properties to the composition while not significantly decreasing the softness of the composition.

According to one aspect of the invention there is provided a composition comprising a soft ethylene elastomer component and an effective amount of filler and compatibilizer to impart improved scratch and mar resistance properties to the composition while not significantly decreasing the softness of the composition.

According to another aspect of the invention there is provided a composition comprising a soft ethylene elastomer component and an effective amount of filler and

silicone to impart improved scratch and mar resistance properties to the composition while not significantly decreasing the softness of the composition.

According to another aspect of the invention there is provided a composition comprising a soft ethylene elastomer component and an effective amount of filler, silicone, and compatibilizer to impart improved scratch and mar resistance properties to the composition while not significantly decreasing the softness of the composition.

According to a further aspect of the invention there is provided a composition comprising a soft ethylene elastomer component and an effective amount of mica and compatibilizer to impart improved scratch and mar resistance properties to the composition while not significantly decreasing the softness of the composition.

According to a further aspect of the invention there is provided a composition comprising a soft ethylene elastomer component and an effective amount of mica and silicone to impart improved scratch and mar resistance properties to the composition while not significantly decreasing the softness of the composition.

According to yet a further aspect of the invention there is provided a composition comprising a soft ethylene elastomer component and an effective amount of mica, compatibilizer and silicone to impart improved scratch and mar resistance properties to the composition while not significantly decreasing the softness of the composition.

Preferred embodiments of the present invention also include a process for preparing the composition and an article of manufacture which has at least one component fabricated from the composition.

The compositions of the present invention have many advantageous properties. In particular they are scratch and mar resistant, while not significantly decreasing the softness of the composition. The composition also results in the production of articles with low gloss.

Detailed Description of the Invention

In a preferred embodiment, the soft ethylene elastomer component used in the present invention is a rheology-modified ethylene elastomer composition comprising from about 50 to about 95% by weight of elastomeric ethylene polymers or EAO polymer blend and from about 5 to about 50% by weight of a high melting polymer, based on the total ethylene elastomer composition.

Elastomeric ethylene polymers that are suitable for use in the ethylene elastomer composition include interpolymers and diene modified interpolymers. Illustrative polymers include ethylene/propylene (EP) copolymers, ethylene/butylene (EB) copolymers, ethylene/octene (EO) copolymers, ethylene/alpha-olefin/diene modified (EAODM) interpolymers and ethylene/propylene/diene modified (EPDM) interpolymers. More specific examples include ultra low linear density polyethylene (ULDPE) such as Attane™ manufactured by The Dow Chemical Company, homogeneously branched linear EAO copolymers such as Tafmer™ manufactured by Mitsui Petrochemicals Company Limited and Exact™ manufactured by Exxon Chemical Company, and homogeneously branched substantially linear EAO polymers such as the Affinity™ polymers available from The Dow Chemical Company and Engage® polymers available from DuPont Dow Elastomers L.L.C.

Preferred EAO polymer blends include homogeneously branched linear and substantially linear ethylene copolymers with a density measured in accordance with ASTM D-792 of 0.85-0.92 g/cc, especially 0.85- 0.90 g/cc and a melt index or I₂ (measured in accordance with ASTM D-1238 (190°C/2.16 kg weight) of 0.01-30 preferably 0.05-10 g/10 min. These substantially linear ethylene copolymers or interpolymers (also known as "SLEPs") are especially preferred.

In addition, the various functionalized ethylene copolymers such as ethylene vinyl acetate (EVA), which contain from 0.5-50 wt % units derived from vinyl acetate, are also suitable. When using an EVA polymer, those that have an I₂ of from 0.01-500, preferably 0.05-50 g/10 min are preferred.

Preferred SLEPs include Engage® polyolefin elastomers and other polymers produced by The Dow Chemical Company and DuPont Dow Elastomers L.L.C.

The high melting polymer useful in the soft ethylene elastomer component is preferably a homopolymer of propylene, a copolymer of propylene with an α -olefin such as ethylene, 1-butene, 1-hexene or 4-methyl-1-pentene. The polymer may also be a blend of a homopolymer and a copolymer, a nucleated homopolymer or a nucleated copolymer. It may further be a nucleated blend of a homopolymer and a copolymer. The α -olefin is preferably ethylene. The copolymer may be a random copolymer or a block copolymer or a blend of a random copolymer and a block copolymer. The high melting polymer is preferably selected from polypropylene homopolymers and polyethylene copolymers with a melt flow rate (MFR) of (230°C and 2.16 kg weight) of 0.3-60 g/10 min, preferably 0.8-40 g/10 min and more preferably 1-35 g/10 min.

A more detailed description of the preferred elastomeric ethylene polymer or EAO polymer blend and high melting polymer is provided in U.S. Patent No. 6,506,842 and WO 98/32795, which are herein incorporated by reference.

The soft ethylene elastomer component is preferably a rheology-modified, composition with low gel levels that enable the composition to maintain true thermoplastic behaviour.

The soft ethylene elastomer component is preferably treated with a suitable organic peroxide in the manner described in U.S. Patent No. 6,506,842 and WO 98/32795. Such suitable organic peroxides used for the purposes of this invention have a half-life of at least one hour at 120° C. Illustrative peroxides include: a series of vulcanizing and polymerization agents that contain α , α' -bis(t-butylperoxy)-diisopropylbenzene and are available from Geo Specialty Chemicals under the trade-mark Vulcup™, a series of agents that contain dicumyl peroxide are available from Geo Specialty Chemicals under the trade-mark Di-cup™, Luperox™ peroxides made by Atofina, North America, or Trigonox™ organic peroxides made by Moury Chemical Company.

Other suitable peroxides include dicumyl peroxide, 2,5-dimethyl-2,5-di-(t-butyl peroxy)hexane, di-t-butylperoxide, 2,5-di(t-amyl peroxy)-2,5-dimethylhexane, 2,5-di-(t-butylperoxy)-2,5-diphenylhexane, bis(alpha-methylbenzyl)peroxide, benzoyl peroxide, t-butyl perbenzoate and bis(t-butylperoxy)-diisopropylbenzene.

The peroxide may also be used with a suitable co-agent such as Type I polar co-agents, which have a relatively low molecular weight. An example of this type of co-agent is trimethylpropane trimethacrylate (TMPTMA) sold by Sartomer under the name SR-350. Type II low polarity co-agent may also be useful such as 1,2-polybutadiene sold by Sartomer under the trade-mark Ricon®. The addition of co-agent may improve the properties of the rheology-modified ethylene elastomer compound, or reduce the amount of peroxide needed to effect the same level of rheology modification.

The preferred ratio of peroxide to co-agent used in this invention is in the range of about 1:1 to about 1:4 peroxide:co-agent, but may vary depending on the particular application.

A more detailed description of the preferred properties of the rheology-modified ethylene elastomer composition is provided in U.S. Patent No. 6,506,842 and WO 98/32795, which are herein incorporated by reference. Most preferably the composition of the present invention has a soft ethylene elastomer component with an STI of between 20 and 110 (calculated using a Kayeness capillary melt rheometer in which the shear rate is the ratio of melt viscosity at 1 sec⁻¹ to that at 1006 sec⁻¹), a MS of ≥ 1.5 , a ST of between 75 and 145 °C (calculated using differential scanning calorimetry to measure the crystallization temperature of the first peak on cool-down) and a UST of between 50 and 160 °C (calculated using thermal mechanical analysis to determine the temperature at which the penetration probe penetrates 600 microns into the sample with the temperature being ramped from room temperature at 3°C per minute).

The soft ethylene elastomer component is combined with an effective amount of filler to improve the scratch and mar resistant properties of the soft ethylene elastomer component. The filler may be a platy filler, such as mica, talc or clay. Alternatively,

the soft ethylene elastomer component may be combined with a hard, spherical, or elongated filler such as activated alumina.

Most preferably, the soft ethylene elastomer component is combined with an effective amount of the filler mica. The mica is preferably a wet-ground mica of 325 mesh. Preferably a muscovite mica is used or alternatively a phlogopite mica.

The filler is present in an amount of from about 1 to about 20 % by weight of the total composition, preferably between from about 2 to about 15% by weight, more preferably from about 3 to about 12% by weight, and further more preferably from about 4 to about 10% by weight.

The filler may also be combined with a compatibilizer to obtain improved scratch and mar resistant properties.

The addition of the filler and compatibilizer into the composition of the present invention may be by direct addition to the soft ethylene elastomer component during compound manufacture or preferably by addition through a pre-made masterbatch. If a masterbatch is used, it is preferably comprised of filler, compatibilizer and base resin in which the compatibilizer comprises from about 1 to about 20% of the total masterbatch composition, while the filler comprises from about 10 to about 90% of the total masterbatch composition. Preferably, the masterbatch comprises from about 5 to about 15% compatibilizer and from about 30 to about 60% filler.

The compatibilizer is preferably a functionalised polyolefin comprising maleic anhydride grafted Engage® or equivalent metallocene polyethylene plastomer, maleic anhydride grafted ethylene propylene copolymer, maleic anhydride grafted ethylene propylene diene monomer, maleic anhydride grafted linear low density polyethylene, maleic anhydride grafted very low density polyethylene or maleic anhydride grafted polypropylene. The compatibilizer acts as a coupling agent in so far as it helps to increase the adhesion of the filler to the soft ethylene elastomer component. The compatibilizer has a polar portion that binds to the filler and a non-polar portion is compatible with the soft ethylene elastomer component to ensure effective adhesion between the filler and the soft ethylene elastomer component. In a preferred

composition of the present invention, the compatibilizer comprises from about 0.1% to about 10%, preferably from about 0.25% to about 7% by weight of the composition.

The base resin in the masterbatch is preferably a carrier resin such as an Engage® polyolefin plastomer, linear low density polyethylene, very low density polyethylene or ethylene propylene copolymer. Where the base resin in the masterbatch is a functionalised resin such as ethylene vinyl acetate, or a copolymer of ethylene and methylacrylic acid or acrylic acid, the functionalised resin may behave in part as a compatibilizer. The base resin in the masterbatch is typically compatible with the elastomeric ethylene polymer of the composition.

Preferably the melt viscosity of the compatibilizer and the base resin that is used in the masterbatch should be similar or somewhat lower than the melt viscosity of the soft ethylene elastomer component to ensure good dispersion.

An example of a mica masterbatch suitable for use in the composition of the present invention would comprise 12% maleic anhydride functionalized resin as a coupling agent (compatibilizer), 30% mica, and a suitable polyethylene carrier resin.

The composition of the present invention and the masterbatch may also comprise additional additives without deviating from the invention as claimed. These additives may include EAOs that have not been rheology modified, process oils, plasticizers, specialty additives and pigments. The specialty additives include flame retardants; antioxidants; surface tension modifiers; anti-block agents; lubricants; antimicrobial agents such as organometallics, isothiazolones, organosulfurs and mercaptans; antioxidants such as phenolics, secondary amines, phosphites and thioesters; antistatic agents such as quaternary ammonium compounds, amines, and ethoxylated, propoxylated or glycerol compounds; hydrolytic stabilizers; lubricants such as fatty acids, fatty alcohols, esters, fatty amides, metallic stearates, paraffinic and microcrystalline waxes, silicones and orthophosphoric acid esters; mold release agents such as fine-particle or powdered solids, soaps, waxes, silicones, polyglycols and complex esters such as trimethylol propane tristearate or pentaerythritol tetrastearate; pigments, dyes and colorants; plasticizers such as esters of dibasic acids (or their

anhydrides) with monohydric alcohols such as o-phthalates, adipates and benzoates; heat stabilizers such as organotin mercaptides, an octyl ester of thioglycolic acid and a barium or cadmium carboxylate; ultraviolet light stabilizers such as a hindered amine, an o-hydroxy-phenylbenzotriazole, a 2-hydroxy,4-alkoxybenzophenone, a salicylate, a cyanoacrylate, a nickel chelate and a benzylidene malonate and oxalanilide; and zeolites, molecular sieves and other known deodorizers. A preferred hindered phenolic antioxidant is Irganox™ 1076 antioxidant, available from Ciba-Geigy Corp.

Additional fillers may also be useful in conjunction with the composition of the present invention. Such fillers may include carbon black, glass, metal carbonates such as calcium carbonate, metal sulfates such as calcium sulfate, talc, clay or graphite fibers. These fillers may impart further scratch and mar resistance properties to the composition.

The additional additives, if used, are typically present in an amount of less than about 45 %, based on the total composition weight. The amount is advantageously from about 0.001 to about 20 %, preferably from about 0.01 to about 15 % and more preferably from about 0.1 to about 10%.

Most preferably a silicone additive is added to the composition to provide enhanced scratch and mar resistant properties. Silicone is added in an amount of 0.10 to 2.50%, preferably 0.20 to 1.00%, most preferably 0.25 to 0.6%. The amount of silicone additive that is added to the composition will be less than the amount of filler in the composition.

The amount of silicone to be added to the composition will depend in part on the fluid viscosity of the silicone to be added. The fluid viscosity is a standard viscosity measured at 25°C. Preferably higher molecular weight silicones, such as polydimethylsiloxane, may be added with a fluid viscosity of at least 1,000 CSt, preferably at least 10,000 CSt. The silicone may be added as a masterbatch or injected as a pure liquid. If in masterbatch form the silicone will typically comprise between about 10% and about 50% of the masterbatch depending on the fluid viscosity of the silicone.

The silicone may be added to the feed throat of the extruder. However, the silicone, which acts as a lubricant in the composition, is preferably added downstream in the extruder to the molten soft ethylene elastomer component.

Where filler is added as a masterbatch, both the masterbatch and the soft ethylene elastomer component may be added to the feed throat of the extruder at the same time. The soft ethylene elastomer component may also be added in its individual components along with the masterbatch. Alternatively, the masterbatch may be added directly to the molten soft ethylene elastomer component after it has melted.

Where filler is added to the soft ethylene elastomer component in its pure form the filler is preferably added after or at least at the same time as the compatibilizer has already been added to the soft ethylene elastomer component although other sequences are possible.

Addition to the feed throat will require that the feed throat be kept at a temperature cold enough to prevent bridging of components in the feed throat. Subsequent processing of the composition occurs by mixing at a temperature of from about 180 to 220°C, preferably from about 190 to about 210°C.

At the end of the extrusion process, the molten polymer extrudate must be cooled to a low temperature before being solidified so that it can be cut into pellets. If the molten polymer extrudate is cut underwater, the water temperature should be maintained at or below about 30 °C, preferably at or below 15 °C.

It will be apparent to those skilled in the art that many types of mixing equipment may be used to make the composition of the present invention including a Farrel Continuous Mixer, single screw or twin screw extruder and buss kneader.

The compositions of the present invention may be formed into articles of manufacture such as parts, sheets, or other forms using any one of a number of conventional procedures for processing elastomer compositions. The compositions can also be drawn into films, multi-layer laminates or extruded sheets, compounded with one or more organic or inorganic substances, injection-molded into articles such as knobs, handles for appliances or consumer goods, used for profile-extruded articles, or used

for coated fabrics or other film and sheet applications. The compositions may be useful in the manufacture of automobile interior parts, automobile exterior parts, consumer goods with soft touch grips and consumer appliances with soft touch surfaces. Other applications may also be possible and are within the knowledge of the person skilled in the art.

The compositions of the present invention have surprisingly improved scratch and mar resistant properties relative to other soft ethylene elastomer compositions. The compositions are also useful in the manufacture of articles with low gloss.

The following examples illustrate, but do not limit the present invention.

Example 1:

All compositions used were made on a twin-screw extruder (38:1 L/D or 50:1 L/D) at a processing temperature of between 190 and 210°C. The throughput was about 150 lbs/hr at a screw speed of 350 RPM. The melt was pelletized using an underwater pelletizer with the cutting water kept cold at about 11-14°C. The mica masterbatch was added to the feed throat. The silicone masterbatch was added part way down the extruder as a side-feed into the molten polymer.

The compositions were formed into embossed sheets using conventional processing means. The embossed sheets had a deep hair cell grain and were pigmented black. The embossed side of each of the sheets was tested for the following properties, which are set out in Table 2:

- (a) shore A hardness of sheet stock under constant load (1 second) was tested according to ASTM D 2240-00;
- (b) % gloss at 60 deg angle using a glossmeter was tested according to ASTM D 523;
- (c) degree of scratching was tested using the 5-finger scratch test described in Ford FLTM BN at 108-13, where a measurement of 5 indicates the most severe scratch line and a measurement of 1 indicates that no visible scratch

lines can be detected on the sheet. Scratch and mar was tested using 1, 2, 3, 6 and 7 Newtons of force.

Of the sheets tested, the "Control" sample is the unmodified composition—called Advantech 8800 A™, commercially available from DuPont. This composition comprises the following components:

- (a) 74.53% by weight of a mixture of low density Engage® resins comprising as the primary component Engage® 8100 and as the secondary component Engage® 8842;
- (b) 22.97% by weight of polypropylene homopolymer resin with a melt flow rate at 230°C, using a 2.1 kg weight, of 12 grams/10 minutes;
- (c) 2.5 % by weight of Vul-Cup® 20P (Geo Specialty Chemicals) comprising 20% organic peroxide/5% silica/75% polypropylene

Sample A comprised a mica masterbatch. The composition of the masterbatch was: 12% maleic anhydride functionalised resin as a coupling agent (compatibilizer), 30% mica, and Engage® 8100. The mica comprised 7 % by weight of the final composition.

Samples B, C and D comprised a mica masterbatch as well as a silicone masterbatch additive. The composition of the samples A, B, C, and D are set out below.

Table 1: Composition of Samples

Sample	Base Component	Additive	% wt additive in final composition
Control	Advantech 8800 A™	None	---
A	Advantech 8800 A™	mica masterbatch	7 % mica
B	Advantech 8800 A™	mica masterbatch and silicone masterbatch	7 % mica, 0.25 % silicone

Sample	Base Component	Additive	% wt additive in final composition
C	Advantech 8800 A™	mica masterbatch and silicone masterbatch	7 % mica, 0.50 % silicone
D	Advantech 8800 A™	mica masterbatch and silicone masterbatch	7% mica, 0.875 % silicone

Table 2: Comparison of properties of the embossed side of extruded sheet stock

Sample	Scratch Test 7N	Scratch Test 6N	Scratch Test 3N	Scratch Test 2N	Scratch Test 1N	Gloss 60°	Shore A Value
Control	3	3	2	1	1	1.2	90.5
A	1	1	1	1	1	0.8	92.5
B	2	1	1	1	1	0.9-1.2	92.6-93.2
C	1	1	1	1	1	0.9-1.1	93.0-93.7
D	1	1	1	1	1	1.0	93.5

Sample A, had the best combined scratch and mar resistance and gloss properties. The softness of the composition was not significantly decreased with the addition of the mica masterbatch in the composition. The addition of silicone also resulted in a composition with improved scratch-and-mar resistance, while maintaining the softness of the composition. Each of the compositions tested, produced a sheet with low gloss.

Example 2:

A sheet comprising Advantech 8800 A™ base resin, 7 % mica and 0.50 % silicone (sample C) was thermoformed into an article, by preheating and forming the sheet into a shape. The embossed side of the sheet was tested against the thermoformed article for scratch and mar resistance using the 5-finger scratch test as described in Ford FLTM BN at 108-13. The % gloss of the embossed side of the sheet was also tested against the thermoformed article at a 60 degree angle using a glossmeter, tested according to ASTM D 523.

Thermoforming was performed using a Sencorp thermoform machine by placing the sheet between a top and bottom platen. The temperature of the top platen was 507 °F

and the temperature of the bottom platen was 580°F. The sheet was pre-heated for 80-120 seconds and then thermoformed for 18 seconds.

Table 3: Comparison of Properties of Thermoformed and Non-Thermoformed Sheet

Sample	Scratch Test 10N	Scratch Test 7N	Scratch Test 5N	Gloss 60°
Control	4	3	2	1.2
Sample C	2-3	1	1	0.7-0.8
Control Thermoformed	4	3	2	1.6
Sample C Thermoformed	1-2	1	1	1.3-1.4

The thermoformed sheet had the same scratch and mar resistance as the non-thermoformed sheet, when tested under a force of 5 and 7 Newtons, and superior scratch and mar resistance properties to the control sheets. At 10 Newtons the thermoformed sheet showed improved scratch and mar resistance over the non-thermoformed sheet and superior results to the control sheets. The gloss of the thermoformed sheet increased slightly when compared with the gloss of the non-thermoformed sheet, but was less than the gloss of the thermoformed control sheet, which did not comprise mica and silicone.

Example 3

Low viscosity injection molded compositions were prepared on a twin-screw extruder (50:1 L/D) at a temperature of between 190 and 210 °C. The throughput was about 150 lbs/hour at a screw speed of 350 rpm. The melt was pelletized using an underwater pelletizer having a water temperature of about 10-14 °C. Mica masterbatch was added to the composition by addition at the feed throat. Silicone masterbatch was also added to the composition by addition part way down the extruder as a side-feed into the molten polymer.

The compositions were injection molded into air-bag doors at a melt temperature of 198-216 °C using a screw speed of 50 rpm and a mold temperature of about 27 °C.

The air bag doors were tested for scratch and mar resistance using the 5 -finger scratch test described in Ford FLTM BN 108-13, where a rating of 5 indicates the most severe scratch line and a measurement of 1 indicates that no visible scratch lines can be observed. Scratch and mar was tested using a 7 Newton force.

The "control" composition was comprised of the following components:

- a) 74.25 % by weight of Engage[®] resin 8130;
- b) 24 % by weight of polypropylene homopolymer (nucleated and comprising antistatic properties) having a melt flow rate of 12 grams/10 minute at 230 °C, using a 2.1 kg weight; and,
- c) 1.75 % by weight of Vul-Cup[®] 20P (Geo Speciality Chemicals) masterbatch comprising 20 % organic peroxide/5% silica/75% polypropylene.

Sample A comprised the "control" composition and 6.8 % weight mica and 0.5 % weight silicone in the final composition. The composition of the mica masterbatch used in Sample A comprised 12 % weight maleic anhydride functionalized resin as a coupling agent (compatibilizer), 30 % weight mica, and Engage[®] 8100.

Table 4: Comparison of the scratch and mar properties of the airbag doors

	Scratch Test 7N (Control)	Scratch Test 7N (Sample A)
Smooth Side	4	2
Grain Side	3	2

The addition of mica and silicone resulted in a significant improvement of the scratch and mar properties compared to the "control" sample.

Various embodiments of the present invention having been thus described in detail by way of example, variations and modifications will be apparent to those skilled in the art. The invention includes all such variations and modifications as fall within the scope of the appended claims.